THE DISTRIBUTION OF ODD NITROGEN IN THE LOWER STRATOSPHERE AND POSSIBLE PERTURBATIONS CAUSED BY STRATOSPHERIC AIR TRANSPORT

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16. Abstract

In the lower stratosphere a significant production of odd nitrogen results from the reaction $N_2O + O(^1D) \rightarrow 2NO$. Since the transport is relatively slow, odd nitrogen builds up with a maximum mixing ratio of 2×10^{-8} at 30 km. Profiles of odd nitrogen, for different latitudes, winter and summer, are computed from one–dimensional transport models. Variations with latitude are small. Horizontal transport is therefore not believed to alter our results significantly.

In order to evaluate the effect of odd nitrogen upon the ozone layer, NO_x profiles are calculated. OH is here a key component, since it converts NO_2 to HNO_3. In the region where ozone is determined by chemistry rather than by transport (above 25 km), NO_2 is found to be relatively abundant.

The effect of stratospheric transport on the NO_x distribution is shown to depend critically upon the height of emission. The effect increases by a factor of 5 or more for a change of flight level from 18 km to 23 km. This strong dependence should be duely considered when future stratospheric transport is discussed.

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THE DISTRIBUTION OF ODD NITROGEN IN THE LOWER STRATOSPHERE AND POSSIBLE PERTURBATIONS CAUSED BY STRATOSPHERIC AIR TRANSPORT

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Abstract

In the lower stratosphere a significant production of odd nitrogen results from the reaction $N_2O + O\,(^1D) \rightarrow 2\,NO$. Since the transport is relatively slow, odd nitrogen builds up with a maximum mixing ratio of 2×10^{-8} at $30\,km$. Profiles of odd nitrogen, for different latitudes, winter and summer, are computed from one-dimensional transport models. Variations with latitude are small. Horizontal transport is therefore not believed to alter our results significantly.

In order to evaluate the effect of odd nitrogen upon the ozone layer, NO_x profiles are calculated. OH is here a key component, since it converts NO_2 to HNO_3. In the region where ozone is determined by chemistry rather than by transport (above $25\,km$), NO_2 is found to be relatively abundant.

The effect of stratospheric transport on the NO_x distribution is shown to depend critically upon the height of emission. The effect increases by a factor of 5 or more for a change of flight level from $18\,km$ to $23\,km$. This strong dependence should be duely considered when future stratospheric transport is discussed.

1.   Introduction

It is well established (Crutzen, 1971, Johnston, 1971, Hesstvedt, 1973), that nitrogen oxides act upon the ozone layer through the reaction

$$NO_2 + O\,(^3P) \rightarrow NO + O_2 \qquad k_1 = 9.2 \times 10^{-12} \tag{1}$$
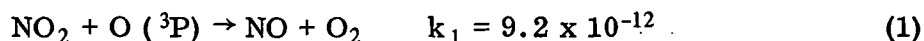
whereby odd oxygen is destroyed. Accordingly, reliable information about the distribution of nitrogen oxides in the lower stratosphere is essential in quantitative assessments of this effect. Production of odd nitrogen takes place in two regions (Crutzen, 1971, Isaksen, 1973). In the upper region, around $100\,km$,

ionic processes result in an important production of NO and N. In the lower region, between 20 and 30 km, NO is produced when N_2O is destroyed by O (^1D). In this region the chemistry appears to be rather complicated. Modeling of the atmosphere with respect to odd nitrogen species, must also comprise a series of oxygen, hydrogen, and carbon species. The model has earlier been presented by Isaksen (1973) and will therefore not be disucssed in detail here. We will, however, discuss production and loss reactions with O (^1D) and OH, which are day components for the nitrogen oxides in the lower stratosphere.

2. The model

Important parameters in the NO_x calculations, such as eddy diffusion coefficient, temperature and the air density, are known to vary markedly with season and latitude. Calculations are therefore made for 60° and 30°, winter, for the Equator, and for 30°, 60°, and 90°, summer. Temperatures and densities are taken from CIRA (1965). The models are one-dimensional, and vertical eddy transport is included. The eddy diffusion coefficients used are given in Figure 1. Up to 27 km the coefficients are taken from Gudiksen et al., (1968). Above 27 km an exponential increase is assumed, as suggested by Lindzen (1971). His indication of a seasonal variation is also adapted.

Hesstvedt (1973) has shown that a two-dimensional model should be considered for computations of the ozone distribution in the lower stratosphere. Since our model is one-dimensional we therefore decided to use observed values of ozone (Hering and Borden, 1964).

Our model is a steady state model, where the photochemistry is averaged over the day. Long lived species which are influenced by eddy diffusion are determined from equations of the type

$$\frac{d\,[X]}{dt} = -\frac{d}{dz}\left(K_z\,[M]\,\frac{d\,\delta_x}{dz}\right) + P_x - Q_x\,[X] = 0 \tag{2}$$

where K_z is the vertical eddy diffusion coefficient, M the total number density, $\delta_x = [X] / [M]$ the mixing ratio of component X, and P_x and $Q_x\,[X]$ are averaged photochemical production and loss terms. This equation is solved by the direct implicit method given by Isaksen (1973), using finite differences.
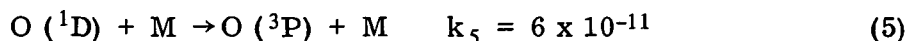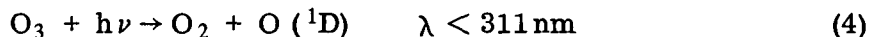
Components with short chemical lifetimes, hours or less, are not influenced by transport processes. The transport term in equation (2) may then be disregarded, and average daytime values are determined from expressions of the type

$$P_x - Q_x\,[X] = 0 \tag{3}$$

where P_x and $Q_x\,[X]$ are average daytime production and loss terms.

3. Atomic oxygen in the excited state and hydroxyl

Two components play an important role in the formation of odd nitrogen species; these components are O (^{1}D) and OH. Odd nitrogen is produced through the reaction (1) which appears to be the only effective source in the stratosphere. Nitrogen oxides will therefore depend strongly on the O (^{1}D) profiles, which are determined from

$$O_3 + h\nu \rightarrow O_2 + O\ (^1D) \qquad \lambda < 311\,nm \qquad (4)$$

$$O\ (^1D) + M \rightarrow O\ (^3P) + M \qquad k_5 = 6\ x\ 10^{-11} \qquad (5)$$

which gives

$$\left[O\ (^1D)\right] = J_3 \cdot \left[O_3\right]/k_5 \cdot \left[M\right] \qquad (6)$$

In our model observed values are used for O_3 and M. J_3 is the daytime average dissociation rate of O_3 for $\lambda < 311\,nm$. Its strong variation with height in the lower stratosphere (Isaksen, 1973) is a result of ozone absorption, and can therefore be calculated from the observed ozone profile.

O (^{1}D) profiles, calculated from equation (6) are given in Figure 2 for different latitudes, summer and winter. The rapid decrease below 50 km is a result of the decrease in J_3. Variations with latitude are small except at high latitudes, winter, where the production is very slow because of the sun.
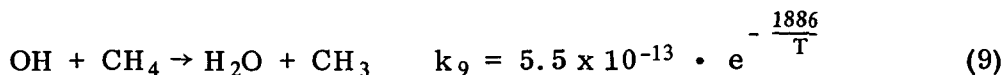
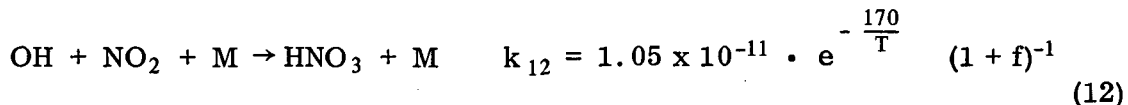Hydroxyl is a second component in the lower stratosphere. It is produced by the reaction

$$NO + HO_2 \rightarrow NO_2 + OH \qquad k_7 = 5\ x\ 10^{-11} \cdot e^{-\frac{1600}{T}} \qquad (7)$$
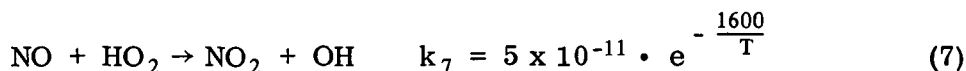
In addition to this reaction there is some production through the reaction

$$O\ (^3P) + HO_2 \rightarrow OH + O_2 \qquad k_8 = 7\ x\ 10^{-11} \qquad (8)$$
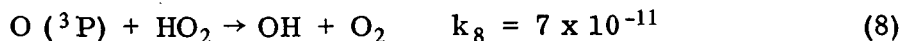
Below 30 km, however, this reaction becomes negligible, since O (^{3}P) decreases rapidly below this height. OH is lost through several reactions. Important reactions in the lower stratosphere are found to be

$$OH + CH_4 \rightarrow H_2O + CH_3 \qquad k_9 = 5.5\ x\ 10^{-13} \cdot e^{-\frac{1886}{T}} \qquad (9)$$

3

$$OH + CO \rightarrow H + CO_2 \qquad k_{10} = 2.1 \times 10^{-13} \cdot e^{-\frac{115}{T}} \qquad (10)$$

$$OH + HNO_3 \rightarrow H_2O + NO_3 \qquad k_{11} = 1.7 \times 10^{-11} \cdot e^{-\frac{1650}{T}} \qquad (11)$$

$$OH + NO_2 + M \rightarrow HNO_3 + M \qquad k_{12} = 1.05 \times 10^{-11} \cdot e^{-\frac{170}{T}} (1 + f)^{-1} \qquad (12)$$
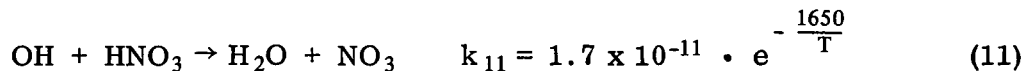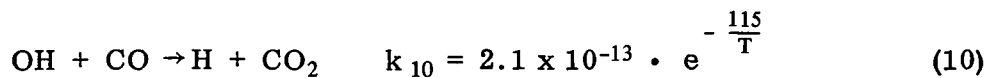
$$f = 1.6 \times 10^{-2} \cdot T^4 / (4 \times 10^{-11} [M])$$

Reactions (9) and (10) are real loss reactions, since CH_3 and H are ultimately converted to HO_2. Reaction (12), on the other hand, is not a real loss since most of OH lost by this reaction is reformed through the reaction

$$HNO_3 + h\nu \rightarrow OH + NO_2 \qquad \lambda < 546 \, nm, \, J_{a \, HNO_3} \qquad (13)$$

The real OH loss is only the fractional part going through reaction (11) and reaction

$$HNO_3 + h\nu \rightarrow HO_2 + NO \qquad \lambda < 370 \, nm, \, J_{b \, HNO_3} \qquad (14)$$
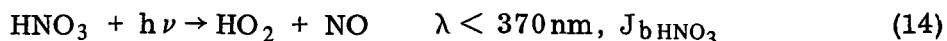
on the short wavelength side of HNO_3 dissociation. It appears that reaction (14) is much less effective than reaction (13). OH may then be determined from

$$[OH] = \frac{(k_7 [NO] + k_8 [O(^3P)]) [HO_2]}{k_9 [CH_4] + K_{10} [CO] + 2k_{11} [HNO_3] + J_{b HNO_3}} \qquad (15)$$

OH can therefore only be determined in a model where all the components occurring in equation (15) are computed simultaneously. This is done in our model. Calculations are repeated until convergence is obtained. The resulting OH profiles are shown in Figure 3. OH has, for all latitudes, a maximum near 45 km with a peak value of $2 - 3 \times 10^{17} cm^{-3}$. Down to the peak level HO_2 is effectively converted to OH through reaction (8), while the destruction through reactions (9), (10), and (12) is slow. Below 45 km this is no longer true and we get a rapid decrease in OH down to the tropopause, where we find a minimum value near $10^5 \, cm^{-3}$. The strong variations with latitude below about 17 km come from the variations in the tropopause level. At the Equator heights below 17 km are in the troposphere, at high latitudes all heights above 8 km are in the stratosphere.
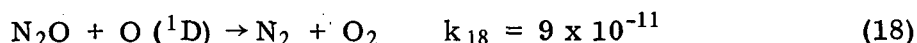
4. Distribution of nitrous oxide

Destruction of N_2O through the reaction

$$N_2O + O(^1D) \rightarrow NO + NO \qquad k_{16} = 9 \times 10^{-11} \qquad (16)$$
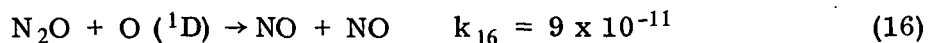
is the only important source of NO_x in the lower stratosphere. Realistic information on the spatial distribution of this component is therefore essential. N_2O has been found to be fairly uniformly distributed in the troposphere with negligible seasonal variations. A mixing ratio of 2.5×10^{-7} is regarded as representative for the troposphere (Schütz et al., 1970). Production of N_2O is expected to be through soil bacteria. So far no photochemical sources are known. It is therefore difficult to estimate production rates. Since the loss rate is small, vertical transport maintain a constant mixing ratio of N_2O in the troposphere, and the above value is used as boundary value at the tropopause. In the lower stratosphere the conditions are quite different. Diffusion is much lower and loss becomes more effective through the reaction

$$N_2O + h\nu \rightarrow N_2 + O \qquad \lambda < 337.5\,nm \qquad (17)$$
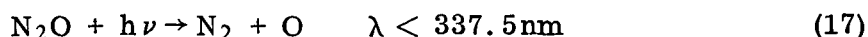
and through the slower reaction

$$N_2O + O(^1D) \rightarrow N_2 + O_2 \qquad k_{18} = 9 \times 10^{-11} \qquad (18)$$

in addition to reaction (16). N_2O profiles are therefore obtained from

$$-\frac{d}{dz}\left(K_z\,[M]\,\frac{d}{dz}\left(\delta_{N_2O}\right)\right) - \left(J_{N_2O} + (k_{16} + k_{18})\left[O(^1D)\right]\right)\,\left[N_2O\right] = 0 \qquad (19)$$

The results are shown in Figure 4. The slope of the profiles depends on the dissociation rates and on the eddy diffusion coefficient. Weak diffusion leads to a rapid decrease in the N_2O mixing ratio $\left(\delta_{N_2O}\right)$. This occurs at 25-30 km, where K_z has a minimum. Variations with latitude and season are relatively small, except for high latitude, winter. Apart from this case, production of NO_x through destruction of N_2O is confined to levels below 35-40 km. At 60°, winter, the low sun makes photodissociation less important, and when in addition the eddy mixing is stronger, N_2O is transported to greater heights before it is dissociated.

## 5.   Production and distribution of odd nitrogen

From the profiles of N_2O and $O(^1D)$, shown in Figures 2 and 4, the production rates of odd nitrogen are easily calculated from

$$P_{NO_y} = 2k_{16} \cdot [O(^1D)] \; [N_2O] \tag{20}$$

The efficiency of this production depends on how efficient reaction (16) is in breaking down N_2O, compared to reactions (17) and (18). Figure 5 shows the loss rate of N_2O from reactions (16) and (17). Loss by reaction (18) is equal to the loss by reaction (16). The curves are for 30° summer. It is clearly demonstrated that reaction (17) is the main loss reaction of N_2O. Only a few percent of the loss will produce nitrogen oxides through reaction (16).

Nitrogen oxide production from eq. (20) is given in Figure 6. In the summer production varies relatively little with latitude. Differences below 20 km is a result of the variation in tropopause level, which influences the $O(^1D)$ densities. At all latitudes, odd nitrogen production increases rapidly up to a marked maximum between 22-26 km. In the winter the production is important over a much wider region. This is clearly demonstrated for 60°, winter, where a substantial production takes place above 40 km, with a maximum around 35 km. The winter profiles are a result of the combined effects of the absorption of radiation producing $O(^1D)$ and the strong eddy diffusion, which effectively transports N_2O to greater heights. At 30°, winter, these effects are less pronounced, with the result that we get a profile more like the summer profiles. Maximum production rates vary from 1.5×10^2 $cm^{-3}s^{-1}$ in the summer hemisphere, to 1.5×10^1 $cm^{-3}s^{-1}$ at 60°, winter. If we add up the total production in a column, the average production for the whole globe becomes 10^8 $cm^{-2}s^{-1}$.

The distribution of total odd nitrogen in the lower stratosphere is easily calculated when its production rate is known. The only photochemical loss of nitrogen oxides in the atmosphere is through the reaction

$$NO + N \rightarrow N_2 + O \qquad k = 2.2 \times 10^{-11} \tag{21}$$
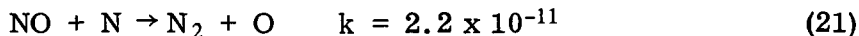
However, atomic nitrogen decreases rapidly below 60 km (Isaksen, 1973). This reaction is therefore of no interest for our problem. Odd nitrogen is therefore removed from the lower stratosphere by vertical eddy diffusion alone. Since K_z increases with height, the particle flux of odd nitrogen, NO_y

$$F = -K_z \, [M] \, \frac{d}{dz} \left(\delta_{NO_y} \right) \tag{22}$$

is directed downwards.

The flux is proportional to the eddy diffusion coefficient and to [M]. It is, therefore clear that the increase in δ_{NO_y}, sufficient to transport away the odd nitrogen produced at a certain height, depends strongly on the heights where the production takes place. Assuming steady state conditions,

$$- \frac{d}{dz} \left(K_z \ [M] \ \frac{d}{dz} \left(\delta_{NO_y} \right) \right) + P_{NO_y} = 0 \qquad (23)$$

gives the height distribution of nitrogen in all odd forms, i.e., in the forms of NO, NO_2, and HNO_3; all other odd components are present in negligible quantities. Results of this calculation are given in Figure 7. As might be expected from the considerations above, there are latitudinal variations in odd nitrogen, due to variations in K_z and in production rates. Above 20 km, where the effect on ozone is of importance, variations with latitude are very small in the summer. The mixing ratio of odd nitrogen increases strongly up to 25 km, the region of the strongest production (Fig. 6). But even above this height there is substantial increase to above 30 km, partly due to the low K_z values. Above 35 km δ_{NO_y} drops as a result of the low mesosphere values used for NO (Isaksen, 1973a). Maximum mixing ratios of 1.2×10^{-8} are found around 32 km.

In the winter the low solar elevation leads to production at higher altitudes. Therefore the increase in mixing ratio takes place at higher altitudes. This is especially true for 60°, winter, where the strong atmospheric absorption gives high mixing ratios in the upper stratosphere. It is, therefore, an increase in odd nitrogen mixing ratios in the whole stratosphere.

6. The distribution of odd nitrogen on NO, NO_2, and HNO_3

In order to estimate the effect on ozone, we shall next see how the total odd oxygen, as computed above, is distributed on the different odd nitrogen species. The initial product is NO through reaction (16). This component will rapidly be converted to NO_2 through reaction

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad k_{24} = 9.3 \times 10^{-13} \exp \left(- \frac{1235}{T} \right) \qquad (24)$$
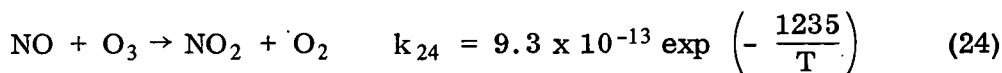
The balance between NO and NO_2 is maintained through solar dissociation of NO_2

$$NO_2 + h\nu \rightarrow NO + O \, (^3P) \qquad 397.5 \, nm \qquad (25)$$
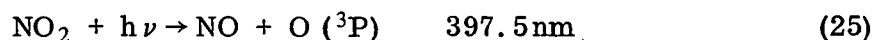
with some contribution from reaction (1).

The conversion to HNO_3 is much slower (reaction (12)). To obtain HNO_3 densities, the loss reactions must be compared with reaction (12). The main loss of HNO_3 is through reactions (13) and (14) with some contribution from reaction (11). NO_3 is rapidly converted to NO and NO_2 through the reactions

$$NO_3 + h\nu \rightarrow NO + O_2 \quad \lambda < 11\mu$$

$$NO_3 + h\nu \rightarrow NO_2 + O \quad \lambda < 700\,nm \tag{26}$$
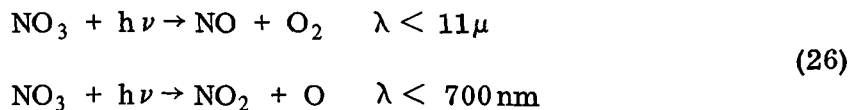
NO_3 is unlikely to be of importance and will be disregarded here.

From the above reactions the distribution of odd nitrogen on NO, NO_2, and HNO_3 can be calculated. Equilibrium between NO and NO_2 is given by reactions (24), (25), and (1). Loss rates of NO and NO_2 are shown in Figure 8 for 30° summer. It is seen that equilibrium is established after a few minutes. Similarly, equilibrium between NO - NO_2 and HNO_3 is given by equations (11) - (14). Equilibrium in this cycle is much slower, as shown in the same figure. Lifetimes decrease from 1 day at 30 km to 10 days at 15 km. The short lifetimes in the NO to NO_2 cycle means that NO is converted to NO_2 a few minutes after sunset, when there is no photodissociation. Likewise, NO is also reproduced rapidly after sunrise from photodissociation of NO_2.

The long lifetimes in the NO - NO_2 - HNO_3 cycle result in negligible HNO_3 variations over the day below 30 km. By confining the equations for equilibrium in the NO - NO_2 and in the NO - NO_2 -HNO_3 cycle, average daytime number densities for NO, NO_2, and HNO_3 are given by

$$[NO] = [NO_y] \frac{J_{HNO_3} + k_{11}[OH]}{J_{HNO_3} + k_{11}[OH] + k'_{12}[OH][M]} \cdot \frac{k_1[O(^3P)] + J_{NO_2}}{k_1[O] + J_{NO_2} + k_{24}[O_3]} \tag{27}$$

$$[NO_2] = [NO_y] \frac{J_{HNO_3} + k_{11}[OH]}{J_{HNO_3} + k_{11}[OH] + k'_{12}[OH][M]} \cdot \frac{k_{24}[O_3]}{k_1[O(^3P)] + J_{NO_2} + k_{24}[O_3]} \tag{28}$$

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$$[HNO_3] = [NO_y] \frac{k'_{12} [OH] [M]}{J_{HNO_3} + k_{11} [OH] + k'_{12} [OH] [M]} \tag{29}$$

k'_{12} gives the efficiency of the coefficient k_{12} in converting $NO + NO_2$ to HNO_3:

$$k'_{12} \approx k_{12} \frac{k_{24} [O_3]}{k_{24} [O_3] + k_1 \cdot [O(^3P)] + J_{NO_2}} \tag{30}$$

The relative abundances of the three nitrogen species, given by equations (27), (28), and (29), are presented in Figure 9 for heights between 10 km and 40 km for 30° summer. HNO_3 is seen to be the dominating component below about 27 km. At 25 km 70% of odd nitrogen present is in the form of HNO_3, and it exceeds 40% at all heights below. The maximum HNO_3 concentrations around 25 km is a result of the high ozone concentration. Below about 15 km HNO_3 increases due to the increase in OH. Above 35 km HNO_3 is present in small quantities. NO_2, which is of special interest in the ozone chemistry, is most abundant around 30 km, where 50% of the total odd nitrogen is in the form of NO_2. Relative densities vary little below 25 km, between 20% and 30%. NO is the most abundant component above 35 km, compared with the other components. Below 30 km it is a minor component. The $NO - NO_2$ cycle is of interest for ozone destruction through reaction (1). As we have already seen, lifetimes are very short. During the day, when ozone is lost through reaction (1), NO and NO_2 are in photochemical equilibrium. When the sum of NO and NO_2 is known, equilibrium values are easily calculated from equations (1), (24), and (25).

NO_x mixing ratios are given in Figure 10 for summer and winter hemisphere. Above 30 km the mixing ratios differ little from odd nitrogen mixing ratios, due to low HNO_3 densities (Fig. 8). Below 25 km, there is a sharp drop down to 10 km, where mixing ratios of about 10^{-9} are obtained for all latitudes. Variations with latitude are very small, except in the winter hemisphere where δ_{NO_x} falls off at higher latitude.

7. Air transport in the stratosphere and its effect on the NO_x distribution

It is evident that nitrogen oxides produced from aircraft flying in the stratosphere will alter the stratospheric NO_x distribution. Since vertical eddy diffusion varies strongly with height, it is of interest to estimate the magnitude of this additional source when the aircraft are assumed to fly at given heights. We shall here consider the effect of a "reference fleet" of 200 four-engine aircraft, each performing 1000 flights of 2-1/2 hours per year. The fuel flow is assumed

to be 5000 kg per hour per engine. The emission of NO (converted to NO_2) is assumed to be 21 g per kg fuel. These 200 aircraft are assumed to fly at the same flight level uniformly routed around the globe. This gives an average production rate of 1.88×10^7 NO_x molecules $cm^{-2}s^{-1}$. Since we use finite differences with steps of 1 km, we have, in our model, a production rate of 1.88×10^2 NO_x molecules $cm^{-2}s^{-1}$. This number will be used as an additional source in equation (23) at given flight levels. The variation in δ_{NO_x}, for flight levels between 15 km and 32 km, is given in Figure 11. The steady state profiles in Figure 10 for 60°, summer, and 30°, winter, are used as a basis, and the increase is given in percent. Since the ozone distribution below 25 km is largely dominated by transport processes (Hesstvedt, 1973 a), the increase in NO_x between 25 km and 30 km is of primary interest for the ozone problem.

As might be expected, the increase in NO_x depends strongly on the flight level. NO_x injection at high altitudes leads to a considerable increase in NO_x. This can be explained by the very fast drop in eddy diffusion coefficient above the tropopause (Fig. 1). The added NO_x has to be transported downwards into the troposphere. Production at heights where K_z is small, therefore means that the gradient in δ_{NO_x} must be much greater than the gradient is at lower heights with higher K_z values, in order to maintain the flux of NO_x given by equation (22).

The increase in δ_{NO_x} is somewhat different in the two diagrams of Figure 11, a result of difference in K_z. In both cases, however, the perturbation in NO_x increases rapidly as the source level, given along the abcissa, is moved upwards. With the source at 18 km, the increase in NO_x is approximately 2% for 60°, summer, and 0.5% for 30°, winter. With the source at 23 km, NO_x increases with 5-10% in the 25-30 km region in both models, and with source at 28 km, NO_x increases by around 20% in both models. The calculations give similar results for the other latitudes. These results have been used in a two-dimensional model to show the effect on the ozone layer (Hesstvedt, 1973 a).

These numbers clearly demonstrate that the effect of stratospheric transport on the NO_x densities depends strongly on the flight levels. Flight levels above 20 km lead to a considerable increase in NO_x. Having in mind that the perturbation (in %) of the ozone layer is roughly one third of the perturbation (in %) of NO_x, the importance of the flight level should be duely considered when the effect of stratospheric transport upon the ozone layer is discussed.

Acknowledgement

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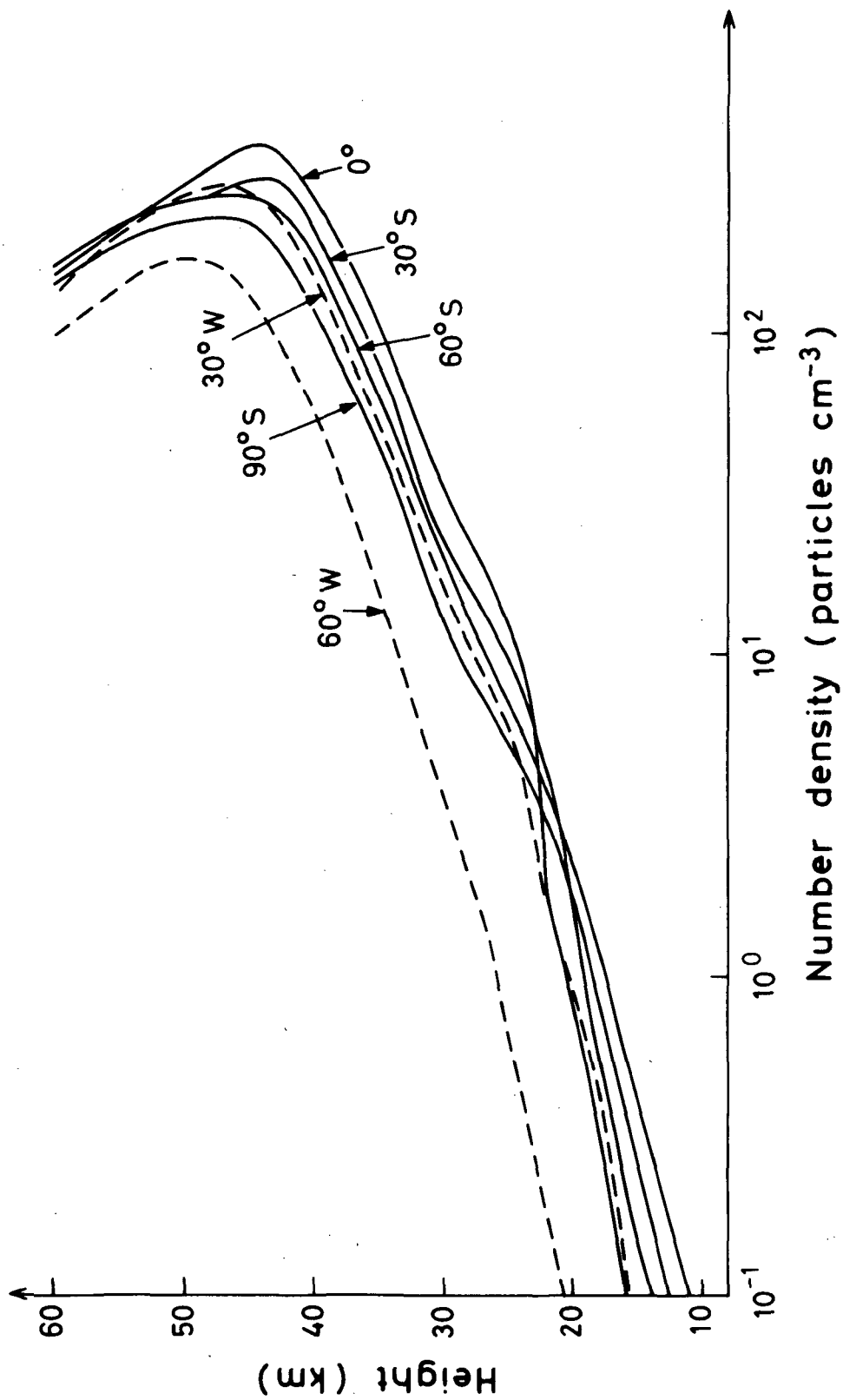
Figure 1. Vertical eddy diffusion coefficient.

13

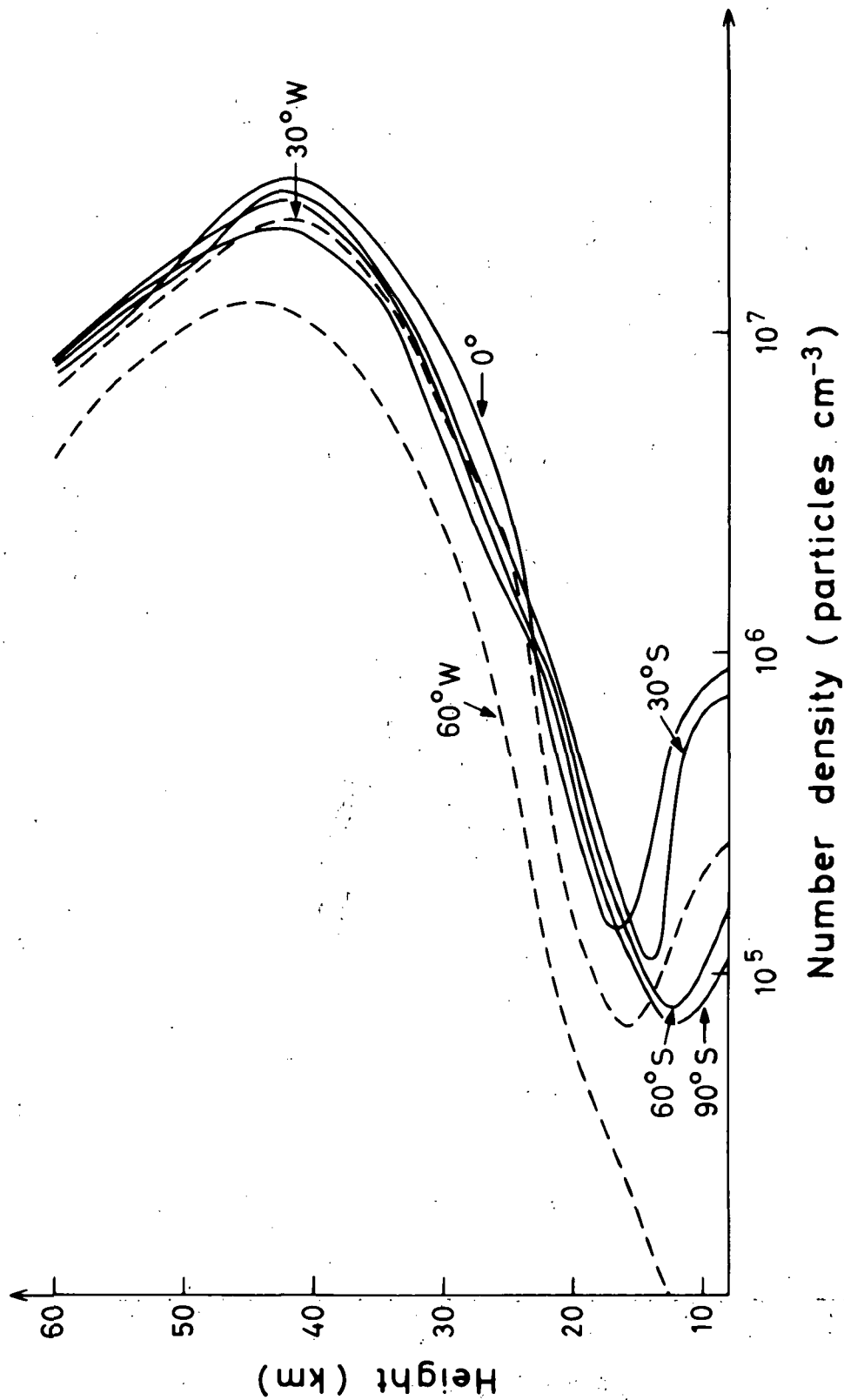Figure 2. Vertical profiles of O(^{1}D) for 90° summer, 60° summer, 30° summer, 0°, 30° winter, and 60° winter.

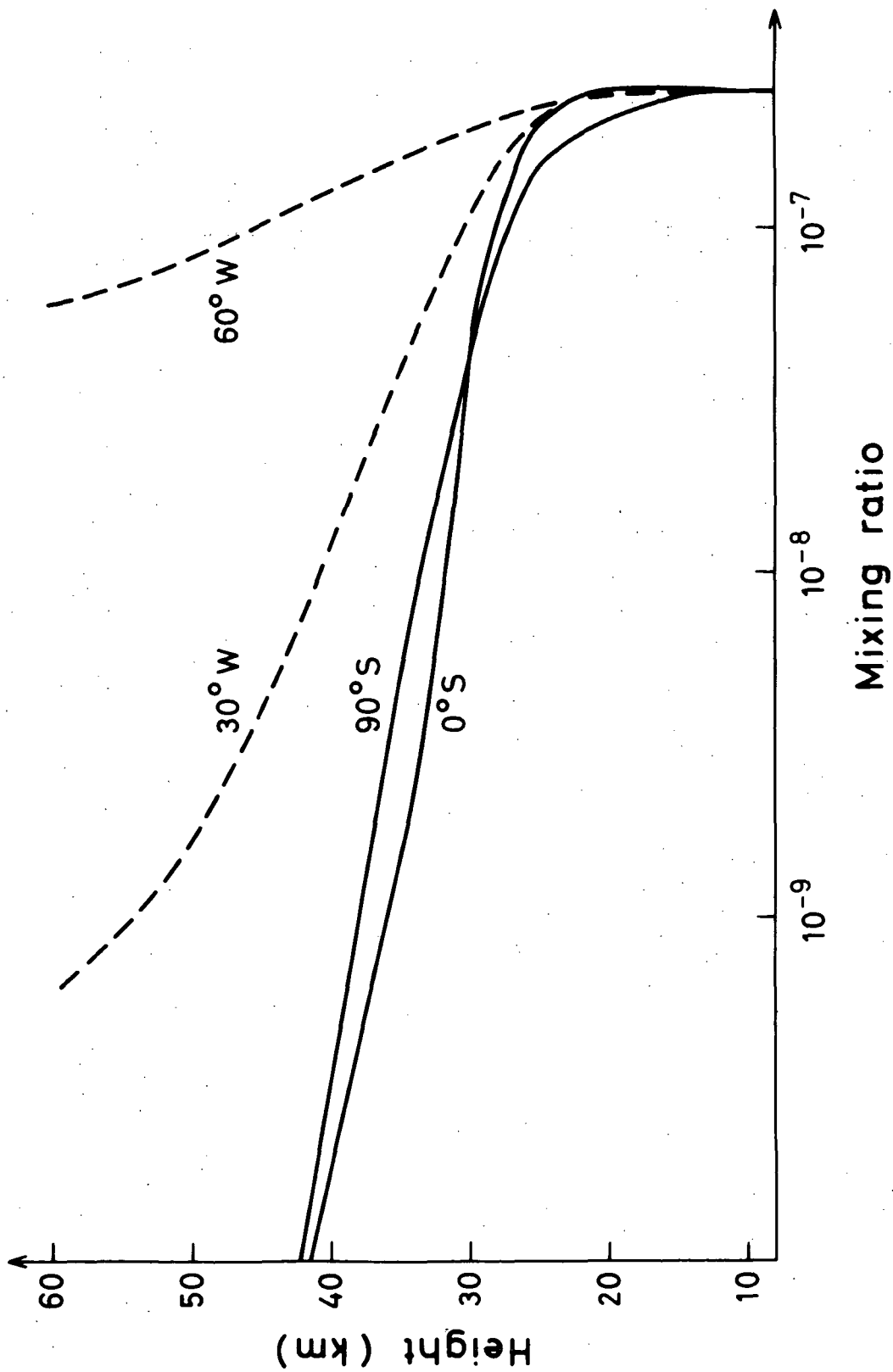Figure 3. Vertical profiles of OH, for the same latitudes as in Figure 2.

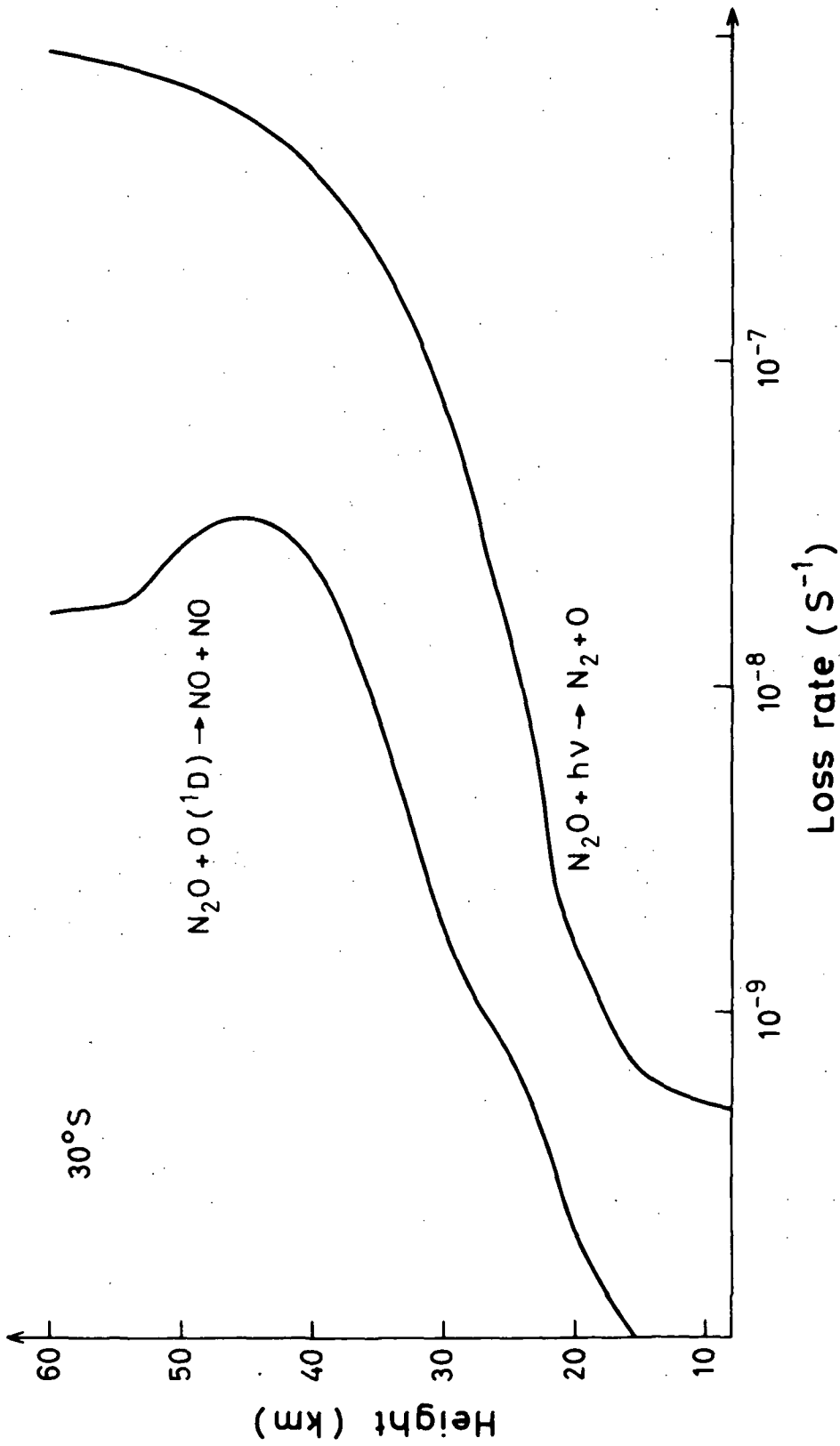Figure 4. Vertical profiles of N_2O for 90° summer, 0°, 30° winter, and 60° winter.

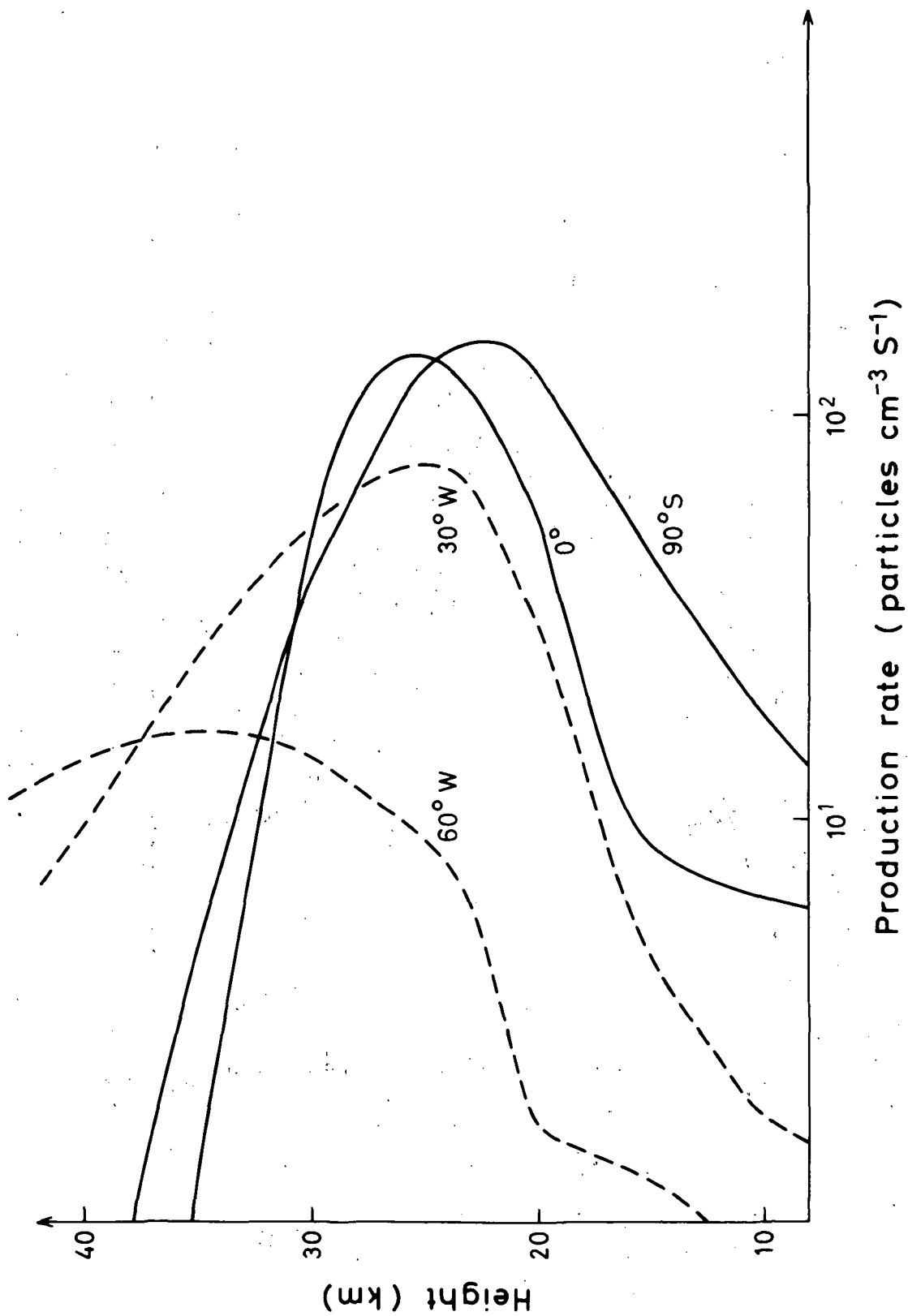Figure 5. Vertical profiles of N_2O loss rates from equations (16) and (18) at 30° summer.

30°S

$N_2O + O(^1D) \rightarrow NO + NO$

$N_2O + h\nu \rightarrow N_2 + O$

Loss rate (S^{-1})

Height (km)

17

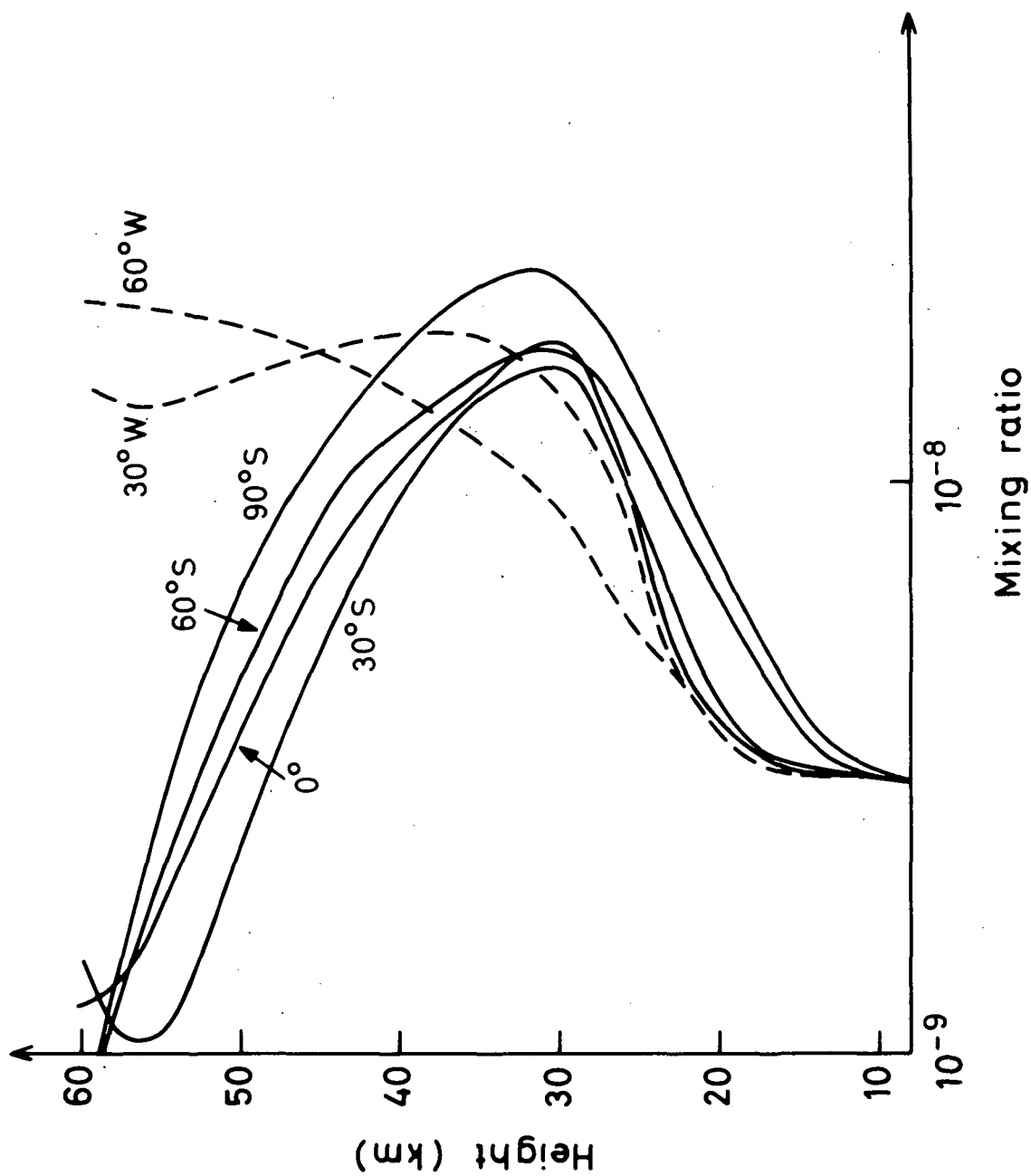Figure 6. Vertical profiles of production rates of odd nitrogen from equation (16) for the same latitudes as in Figure 4.

Figure 7. Profiles of odd nitrogen mixing ratios for the same latitudes as in Figure 2.
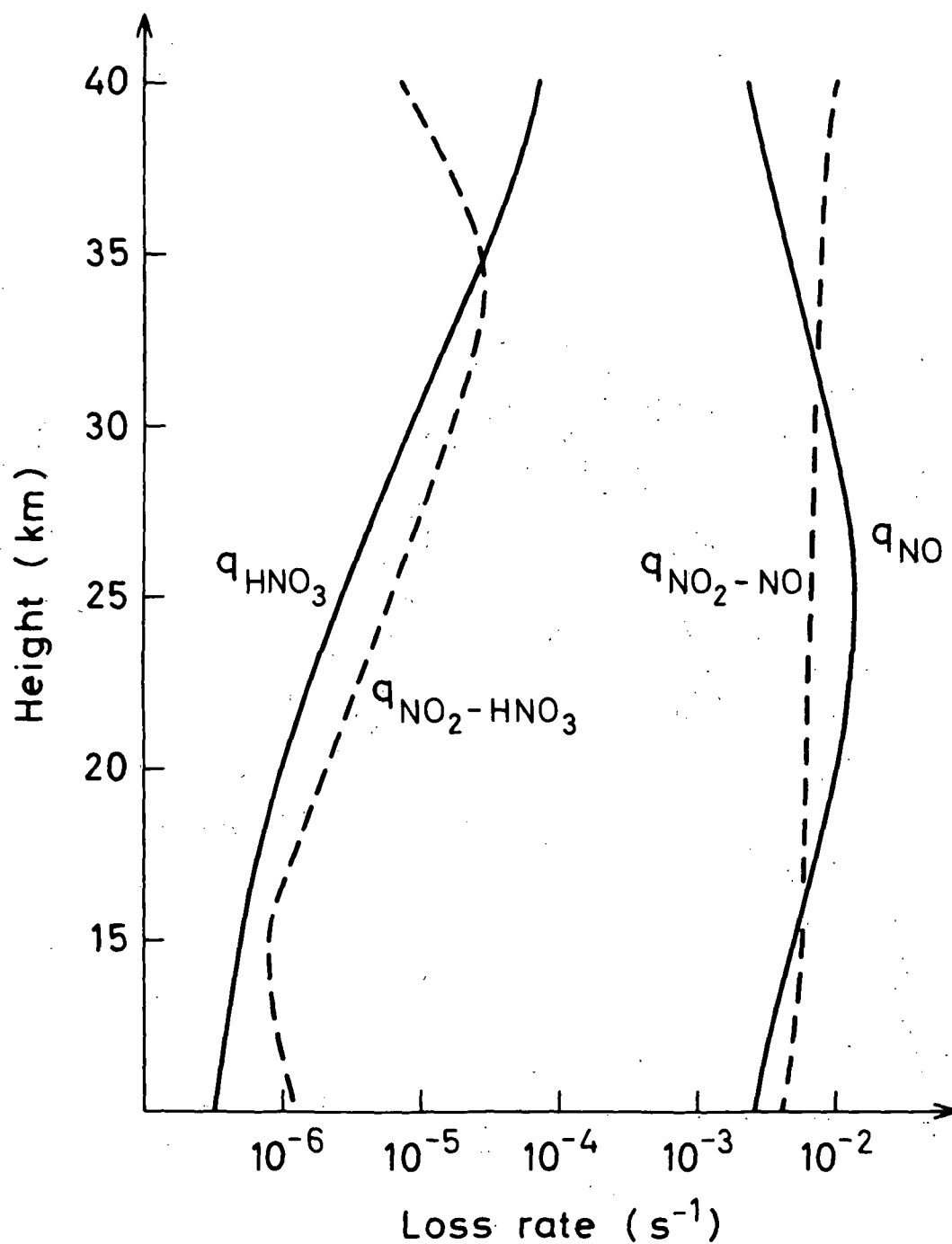
Figure 8. Vertical profiles of odd nitrogen loss rates in the NO–NO$_2$ cycle (curves q_{NO_2-NO}, and q_{NO}) and in the NO–NO$_2$–HNO$_3$ cycle (curves $q_{NO_2-HNO_3}$ and q_{HNO_3}) for 30° summer.
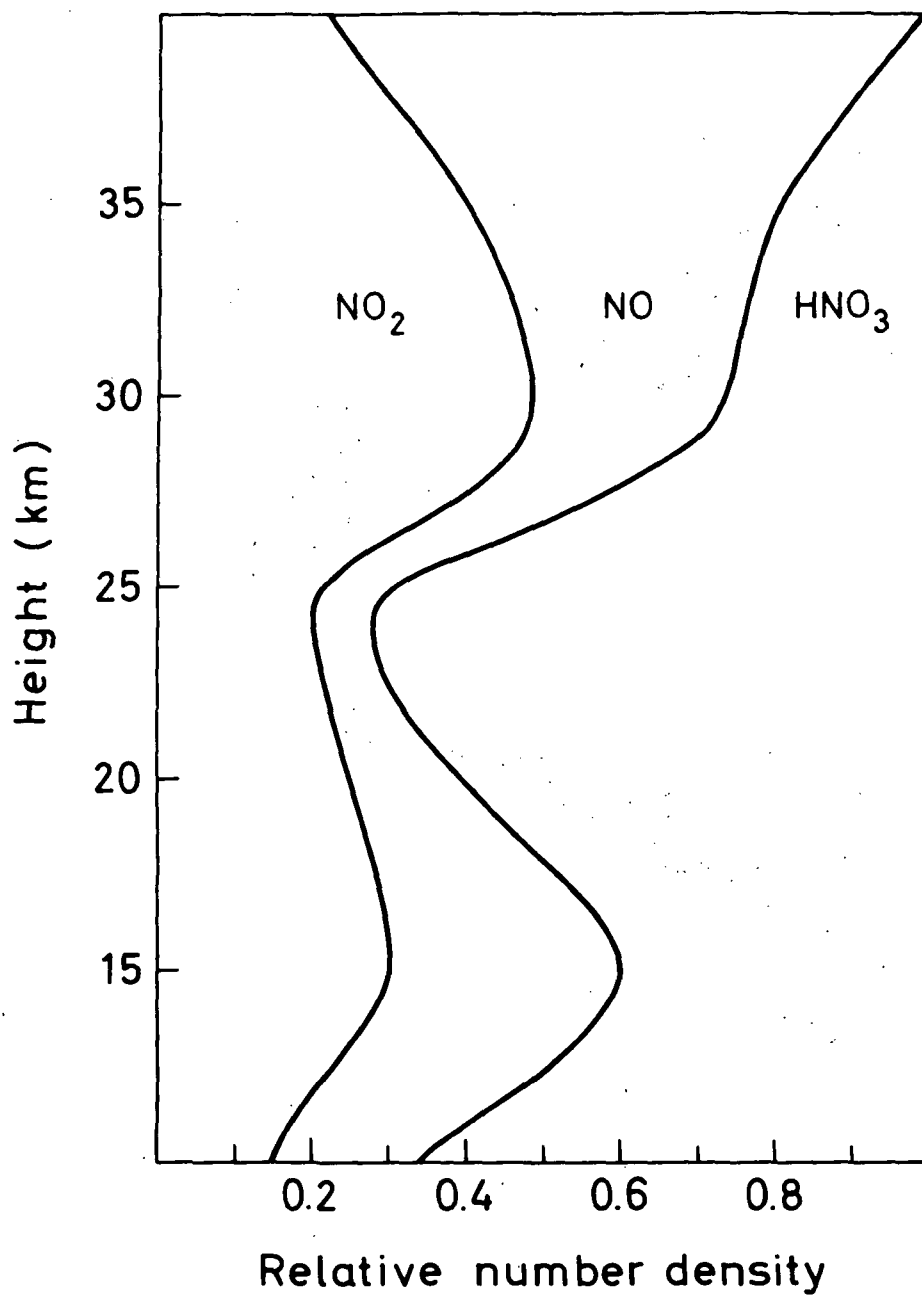
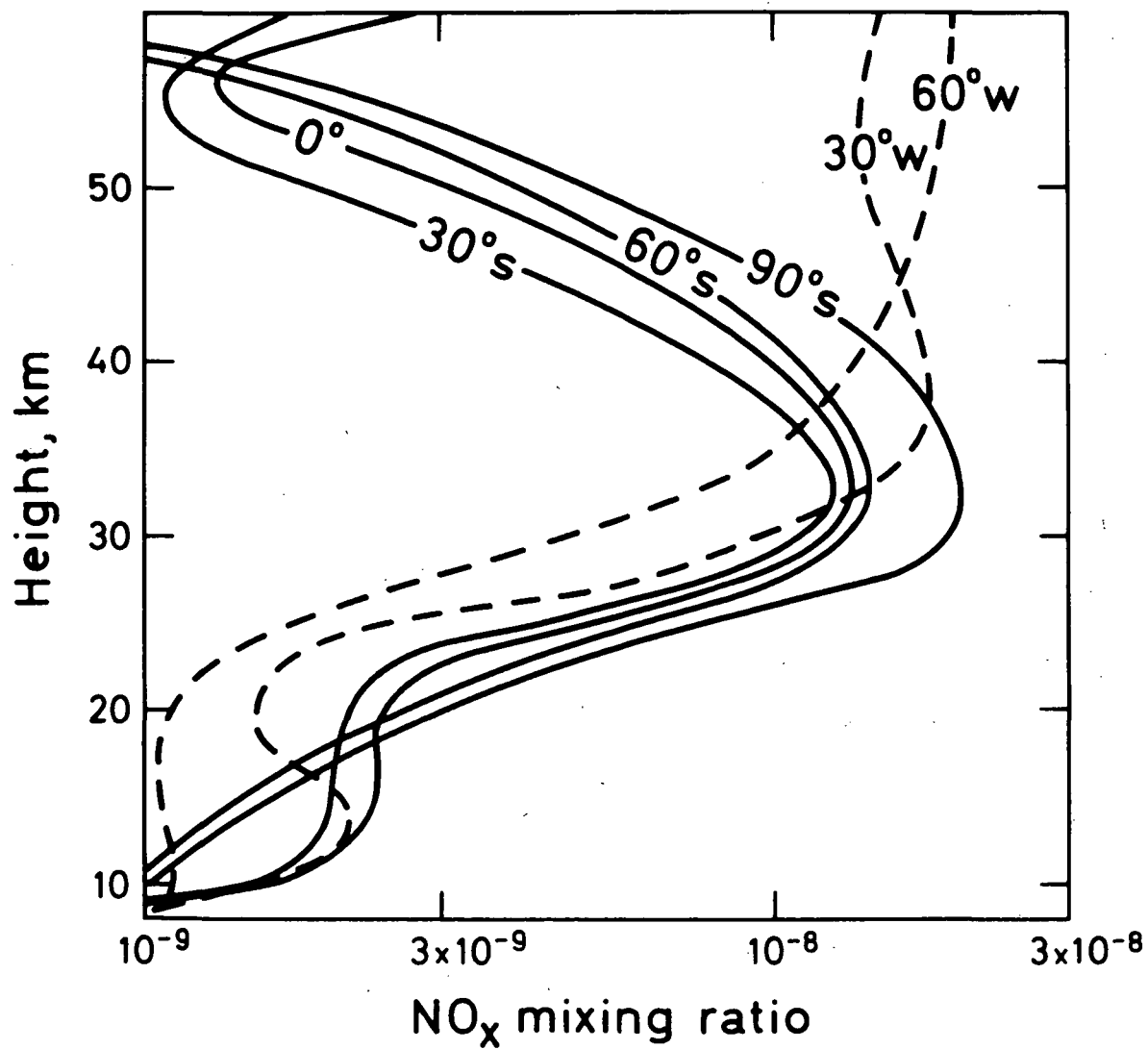Figure 9. Distribution of odd nitrogen on NO, NO$_2$, and HNO$_3$.

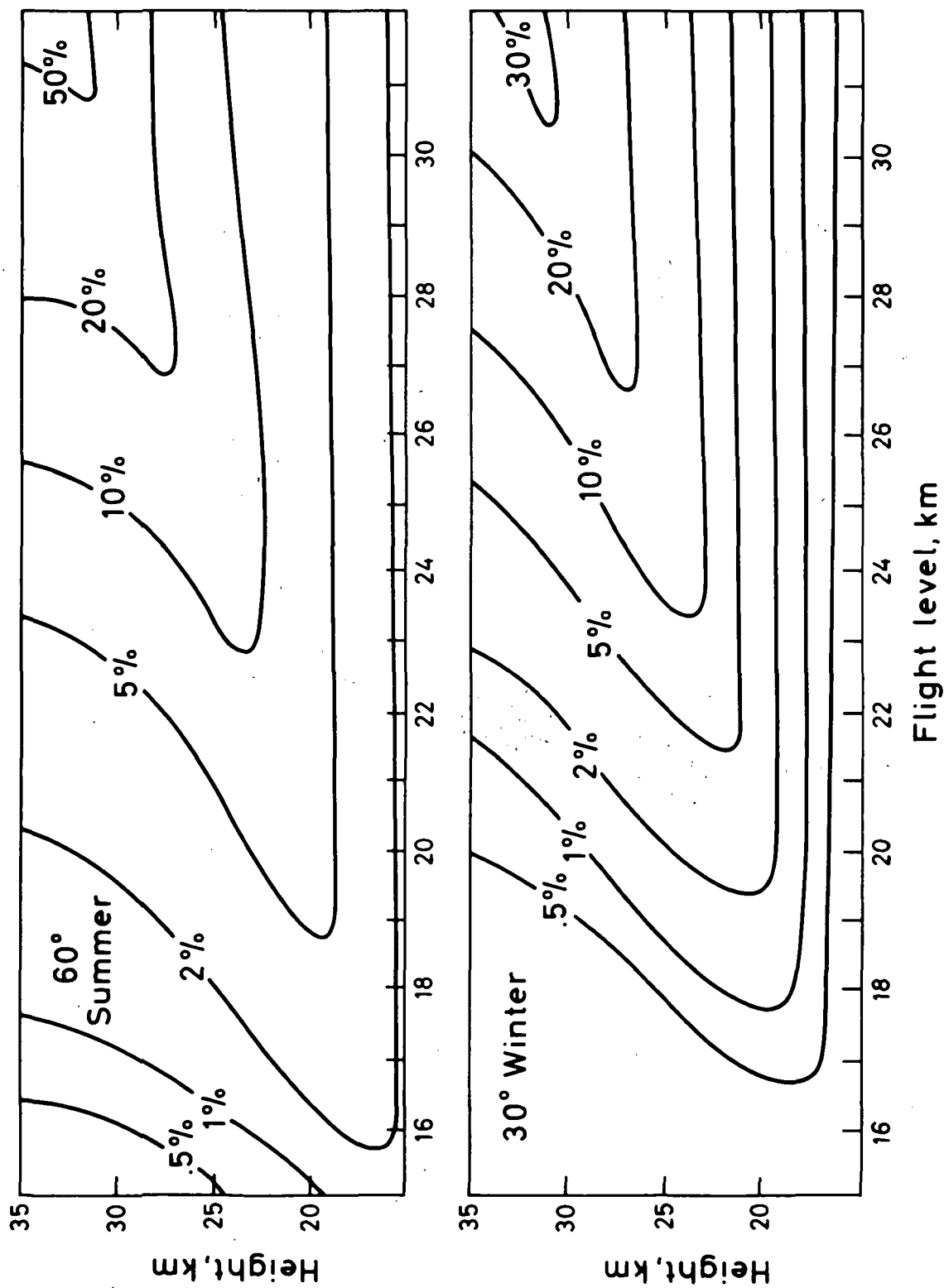Figure 10. Vertical profiles of NO_x mixing ratio.

Figure 11. Increase (in %) of NO_x mixing ratio caused by a fleet of 200 SST aircraft as a function of flight level (horizontal axis).

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